

**REMARKS**

Claims 1-16 are pending in the application.

Claims 14 and 16 have been amended to recite that the molar ratio of  $\text{LiPF}_6$  to  $\text{LiBF}_4$  is 1:9 to 9:1. This amendment is supported in the specification at least in Example 5. No new matter has been added by this amendment.

In the present Office Action, the Examiner has rejected claims 1-3, 5, 12, 14, and 16 under 35 U.S.C. § 102(e) as being anticipated by U.S. Patent Application Publication No. 2002/0064712 of Sekino et al. ("Sekino"). Further, claims 1-9 and 12-16 have been rejected under 35 U.S.C. § 103(a) as being unpatentable over U.S. Patent Application Publication No. 2003/0118913 of Takami et al. ("Takami") in view of U.S. Patent No. 6,315,918 of Mita et al. ("Mita"). The Examiner has rejected claims 10-11 under 35 U.S.C. § 103(a) as being unpatentable over Takami in view of Mita and further in view of EP 0796510 ("EP '510"). Finally, claim 15 has been rejected under 35 U.S.C. § 103(a) as being unpatentable over Takami in view of Mita and further in view of U.S. Patent Application Publication No. 2002/0001756 of Hamamoto et al. ("Hamamoto"). Applicants respectfully traverse these rejections and the arguments in support thereof, and respectfully request reconsideration and withdrawal of the rejections.

**Rejection Under § 102(e) Based on Sekino**

The Examiner argues that claims 1-3, 5, 12, 14, and 16 are anticipated by Sekino. However, the filing date of Sekino, September 24, 2001, is subsequent to the filing date (January 29, 2001) of one of the priority documents of the present application, JP 2001-019556. Submitted herewith is a verified translation of JP 2001-019556. Since rejected claims 1-3, 5, and 12 are fully supported in the priority document, Sekino is not prior art against at least these claims.

Regarding claims 14 and 16, which recite that the molar ratio of  $\text{LiPF}_6$  to  $\text{LiBF}_4$  is 1:9 to 9:1, the Examiner argues that Sekino teaches in Section 0110 the use of  $\text{LiPF}_6$  and  $\text{LiBF}_4$  as a single or a plurality of solutes. However, the claimed ratio of  $\text{LiPF}_6$  to  $\text{LiBF}_4$  is not taught or suggested. Therefore, for these reasons, reconsideration and withdrawal of the § 102(e) rejection based on Sekino are respectfully requested. Further, any rejection of present claims 14 and 16 over Sekino would have to be based on U.S.C. § 103(a). As demonstrated below, the criticality

of this molar ratio would not be expected based on Sekino, so that a rejection under § 103(a) would not be proper.

*Rejection Under 103(a) Based on Takami in view of Mita*

Regarding claims 1-9 and 12, the Examiner argues that Takami teaches a non-aqueous electrolyte secondary battery comprising an electrode group containing a positive electrode, a negative electrode and a non-aqueous electrolyte including solvents and lithium salts dissolved therein. It is allegedly disclosed to use cyclic carbonates, such as ethylene carbonate, propylene carbonate, or vinylcarbonate, together with  $\gamma$ -butyrolactone, and further to be desirable to prepare a mixed solvent including an aromatic compound. Various preferred combinations of non-aqueous solvents are allegedly taught, particularly in paragraphs [0057]-[0059] and in Examples 15-18.

Regarding claims 12-13, the Examiner argues that Takami discloses the claimed positive and negative electrode materials and the employment of lithium salts, such as  $\text{LiPF}_6$  and  $\text{LiBF}_4$ , as recited in claims 14 and 16. Finally, the Examiner argues that Takami teaches in paragraph 63 a solvent containing a benzene-like or derivative compound, as well as a specific solvent mixing solution in Example 14. For these reasons, the Examiner concludes that Takami discloses a non-aqueous electrolyte secondary battery. The Examiner acknowledges that Takami does not disclose the specific use of vinylethylene carbonate (VEC) solvent as claimed.

However, the Examiner argues that Mita discloses non-aqueous electrolytic solutions employed in the electrochemical field and specifically teaches that organic solvent-based electrolytic solutions (non-aqueous electrolytic solutions) show a high potential window of electrochemical stability. Mita allegedly discusses secondary batteries. The Examiner further argues that Mita teaches that cyclic carbonic esters can be suitably used as a solvent for the electrolytic solutions used in batteries; the solvents may be used singly or in combination and include ethylene carbonate, butylene carbonates, vinylene carbonate, and cyclic esters such as  $\gamma$ -butyrolactone, 3-methyl-  $\gamma$ -butyrolactone and 2-methyl- $\gamma$ -butyrolactone. In particular, Mita allegedly exemplifies the use of VEC in a mixed solvent.

Accordingly, the Examiner concludes that it would have been obvious to one having ordinary skill in the art at the time of the invention to use the VEC solvent of Mita in the secondary battery of Takami because Mita teaches that the cyclic carbonic esters can be suitably

used as a solvent for the electrolytic solutions used in batteries, since such solvents show excellent acid resistance, are not oxidized when allowed to stand in the atmosphere, and are chemically stable without reacting with water under normal storage conditions or reacting with highly reactive substances such as metal lithium. In addition, such cyclic carbonic esters are allegedly physically safe, not readily thermally decomposed, and are flame-retardant and resistant to electrochemical oxidation or reduction. Furthermore, the Examiner notes that Mita and Takami share the same field of endeavor of providing suitable non-aqueous electrolytic solutions for electrochemical applications or resistance to electrochemical oxidation-reduction. Therefore, the Examiner concludes that both references are relevant to each other. Finally, since Takami allegedly teaches the use of cyclic carbonic acid esters having at least one carbon-carbon unsaturated bond in a combination of mixed solvents, the Examiner concludes that one cyclic carbonic acid ester can be used interchangeably to substitute for another because they are deemed to be functionally and chemically equivalent. Applicants respectfully traverse this reaction as follows.

In paragraphs [0057] to [0059], Takami teaches a mixed non-aqueous solvent which contains  $\gamma$ -butyrolactone as a main component, combined with a cyclic carbonate such as propylene carbonate, ethylene carbonate, or vinylene carbonate. However, Takami does not teach that the solvent may comprise VEC, or that the solvent must contain VEC in addition to a cyclic carboxylic acid ester and a cyclic carbonic ester having no carbon-carbon unsaturated bond as claimed.

In contrast, Mita is directed to a non-aqueous electrolytic solution for capacitors. According to Mita, VEC is used as one of the main solvents for the electrolyte, which is not decomposed during charge and discharge. There is no teaching or suggestion in Mita that VEC forms a coating on the surface of the negative electrode of a non-aqueous electrolyte secondary battery as occurs in the present invention.

The present invention is directed to a non-aqueous electrolyte secondary battery which contains a non-aqueous solvent containing (A) a cyclic carboxylic acid ester; (B) a cyclic carbonic acid ester having at least one carbon-carbon unsaturated bond; and (C) a cyclic carbonic acid ester having no carbon-carbon unsaturated bond. Component (B) comprises vinylene carbonate (VEC). VEC is an important component of the non-aqueous electrolyte and is decomposed by reduction faster than the solvent at a slightly more noble potential than 0 V

(Li/Li<sup>+</sup>). Since the unsaturated bond is prone to polymerization, VEC undergoes a chain reaction on the negative electrode and rapidly forms a close and strong film on the negative electrode surface. This film serves as a physical barrier which inhibits the contact of solvent molecules around lithium ions with the negative electrode, thereby suppressing the reductive decomposition of the cyclic carboxylic acid ester on the negative electrode (see page 5, lines 18-25).

Example 6 and Table 12 of the present application describe and demonstrate the preparation of a battery according to claim 1, in which the non-aqueous electrolyte contains 2% VC and 1% VEC (1G) and a comparative battery (2G) which contains only 2% VC in the non-aqueous electrolyte solution. As shown in Table 13, battery 1G generated less gas, 1.9 ml compared with 3.0 ml. These results demonstrate that using VC and VEC together is an effective way to reduce the amount of gas generated after the battery cycles. Such a result would not be expected based on Takami (which does not teach or suggest VEC), Mita (which does not teach or suggest VEC in combination with VC) or their combination, and thus would overcome any *prima facie* case of obviousness were one to be established.

The Examiner argues that these examples employ specific non-aqueous electrolytic solutions in restricted battery embodiments including, for instance (i) specific organic solvent compounds; (ii) specific solvent compositions and molar amounts; (iii) specific battery components, active materials, and their constituents such as negative and positive electrodes comprising respective active materials, binders, conductive agents, current collectors, etc., and battery separators, gaskets, and the like; (iv) specific battery structural arrangements; (v) specific battery dimensions and the like. Therefore, the Examiner concludes that the battery embodiments from which the argued unexpected results derive are strictly related to very specific battery exemplifications that are not commensurate to the claimed invention.

Applicants respectfully traverse the Examiner's conclusion. The results described in Example 6 and shown in Tables 12 and 13 demonstrate the effect of including VEC in a battery according to the present invention. Specifically, battery 1G includes components (A), (B), and (C), including VEC, and is thus commensurate with claim 1. In contrast, battery 2G is a comparative battery which does not contain VEC, and it can be clearly seen that there is a difference in the amount of gas generated, which would not be expected based on the proposed combination of Takami and Mita.

Claims 14 and 16 recite that the molar ratio of  $\text{LiPF}_6$  to  $\text{LiBF}_4$  present in the solute is 1:9 to 9:1. As previously explained, by using a non-aqueous electrolyte containing this solute in a non-aqueous electrolyte secondary battery according to the invention, battery cycle characteristics are improved (see page 15, line 26 to page 16, line 1). As taught in the present application, while  $\text{LiPF}_6$  has excellent electrical characteristics, it is chemically unstable since it produces hydrofluoric acid. Cyclic carboxylic acid esters (A) are prone to decomposition by hydrofluoric acid, possibly deteriorating the cycle characteristics of the battery. Additionally, the use of  $\text{LiBF}_4$  also deteriorates cycle characteristics of the battery because the degree of electrolytic dissociation of  $\text{LiBF}_4$  is small or a film formed on the surface of the negative electrode comprising graphite is weak. However, as shown in Tables 10 and 11, the use of both salts in combination, particularly in the claimed ratio, as demonstrated below, improves cycle characteristics of the battery and decreases the amount of gas generated, and also results in favorable capacity maintenance rates and cycle life.

As described in Example 5 of the present application and in the Declaration of Atsushi Ueda Under 37 C.F.R. § 1.132, filed herewith ("the Ueda Declaration"), capacity maintenance rates after storage, cycle lives, and the amounts of gas generated after cycles were examined for a series of batteries in which the mixing molar ratio of  $\text{LiPF}_6$  to  $\text{LiBF}_4$  was varied while the total concentration of  $\text{LiPF}_6$  and  $\text{LiBF}_4$  was kept constant. As shown in the Table of the Ueda Declaration, batteries 2F and 3F, for example, which included both  $\text{LiPF}_6$  and  $\text{LiBF}_4$  in the solute, exhibited an improvement in cycle life and a decrease in the amount of generated gas, both favorable and desirable results. In contrast, none of the cited prior art teaches or suggests a solute containing both  $\text{LiPF}_6$  and  $\text{LiBF}_4$ . Further, none of the prior art suggests the unexpected and unique results which have been obtained by Applicants by including the combination of the two lithium salts in the non-aqueous electrolyte.

The criticality of the ratio between  $\text{LiPF}_6$  and  $\text{LiBF}_4$  can be seen by considering the data presented in the Table of the Ueda Declaration and described in paragraphs 14-16. When the relative amount of  $\text{LiPF}_6$  is high (see batteries 2F and 3F, for example), the capacity maintenance rate after storage at high temperature is high and the amount of gas generated after the cycles is low. However, when the relative amount of  $\text{LiPF}_6$  is greater than 0.9 M (ratio of  $\text{LiPF}_6$ : $\text{LiBF}_4$  greater than 9:1), as in batteries (a) and 1F, the cycle life is unfavorably reduced to 320 or 300, respectively.

On the other hand, as the relative amount of  $\text{LiBF}_4$  is increased, the capacity maintenance rate tends to decrease and the amount of gas generated after the cycles tends to increase. When the relative amount of  $\text{LiBF}_4$  exceeds 0.9 M (ratio of  $\text{LiPF}_6$ : $\text{LiBF}_4$  less than 1:9), as in batteries (b) and 5F, the capacity maintenance rate becomes as small as 81% and the amount of gas generated after the cycles increases to as large as 2.8 or 3.0 mL, respectively.

It can thus be concluded from the Table that when the  $\text{LiPF}_6$ : $\text{LiBF}_4$  molar ratio is maintained between 9:1 and 1:9, the resulting batteries exhibit excellent capacity maintenance rates and cycle lives, and small amounts of gas are generated after the cycles. However, when the molar ratio falls outside this range, inferior results are obtained.

As described in paragraph 17 of the Ueda Declaration, it can be expected that a battery which contains VEC in the non-aqueous solvent, as well as  $\text{LiPF}_6$  and  $\text{LiBF}_4$  in the solute in a molar ratio of 9:1 to 1:9 (as recited in claim 14), will exhibit similar favorable properties to those exemplified by the batteries described in the Declaration.

Applicants have thus found that not only does the combination of the two lithium salts result in favorable battery properties; but have also determined, via experimentation, the critical ratio of these components for optimum battery performance. Since neither Takami nor Mita teaches or suggests the combination of these salts, nor the beneficial properties obtained by such a combination, the criticality of the relative ratio is certainly not expected based on these references. Accordingly, even if the combination of Takami and Mita were proper, it would not teach all of the elements of these claims.

For all of these reasons, reconsideration and withdrawal of the § 103(a) rejection are respectfully requested.

*Rejection Under § 103(a) Based on Takami in view of Mita and EP '510*

Regarding claims 10-11, the Examiner acknowledges that even the combination of Takami and Mita would not teach that the solvent comprises a glime. However, EP '510 allegedly discloses a non-aqueous electrolyte system consisting of a solvent mixture containing ethylene carbonate,  $\gamma$ -valerolactone and optionally containing one or more additional solvents selected from other organic carbonates such as glymes. Therefore, the Examiner concludes that in view of the teachings of EP '510, it would have been obvious to one skilled in the art at the

time of the invention to use the solvent comprising a glime of EP '510 in the solvent mixture of Takami/Mita. Applicants respectfully traverse this rejection as follows.

As previously explained, even if the proposed combination of Takami and Mita were valid, Applicants' invention demonstrates unexpected results which would overcome any *prima facie* case of obviousness were one to be established. Specifically, it would not be expected that inclusion of VEC would reduce the amount of gas generated, as exemplified by the present invention. EP '510 also does not suggest these results, and accordingly, reconsideration and withdrawal of the §103(a) rejection are respectfully requested.

Rejection Under §103(a) Based on Takami in view of Mita and in view of Hamamoto

The Examiner acknowledges that the proposed combination of Takami and Mita does not disclose that the solvent comprises a derivative of benzene. However, Hamamoto allegedly teaches in the abstract that a non-aqueous electrolytic solution which may be used for a lithium secondary battery employs a non-aqueous electrolytic solution which comprises a non-aqueous solvent and an electrolyte which contains a biphenyl derivative. In view of these teachings, the Examiner concludes that it would have been obvious to one having ordinary skill in the art at the time of the invention to use the solvent of Hamamoto comprising a derivative of benzene in the solvent mixture of Takami and Mita, as Hamamoto teaches that by using the benzene derivative as a solvent, a non-aqueous electrolytic solution which is favorably employable for a lithium secondary battery and which shows high battery performance, such as high electric capacity and high cycling performance, under maximum operation voltage condition or elevated temperature is obtained. Applicants respectfully traverse this rejection as follows.

As previously explained, even if the proposed combination of Takami and Mita were valid, Applicants' invention demonstrates unexpected results which would overcome any *prima facie* case of obviousness were one to be established. Hamamoto EP '510 also does not suggest these results, since Hamamoto does not teach or suggest the claimed non-aqueous solvent containing a cyclic carbonic ester which contains VEC. In particular, Hamamoto in paragraph [0018] teaches that the non-aqueous solvent preferably comprises a combination of a cyclic carbonate and a linear chain carbonate, a high dielectric constant solvent such as ethylene carbonate, propylene carbonate, or butylene carbonate, and a low viscosity solvent, such as  $\gamma$ -butyrolactone or dimethyl carbonate, for example. However, Hamamoto does not teach or

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suggest VEC, nor a solvent which contains, in addition to VEC, a category (A) and a category (C) component. Accordingly, in view of the unexpected results of Applicants' invention, reconsideration and withdrawal of the §103(a) rejection are respectfully requested.

In view of the preceding amendments and remarks, Applicants respectfully submit that all of the pending claims are patentably distinct over the prior art of record and in condition for allowance. A Notice of Allowance is respectfully requested.

Respectfully submitted,

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Encl: Request for Continued Examination  
Transmittal of Verified Translation of Priority Document  
Verified Translation of JP 2001-019556  
Petition for Extension of Time (one month)  
Declaration of Atsushi Ueda Under 37 C.F.R. § 1.132